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High Pressure Windowed Chamber Burn Rate Determination of Liquid Propellant XM46

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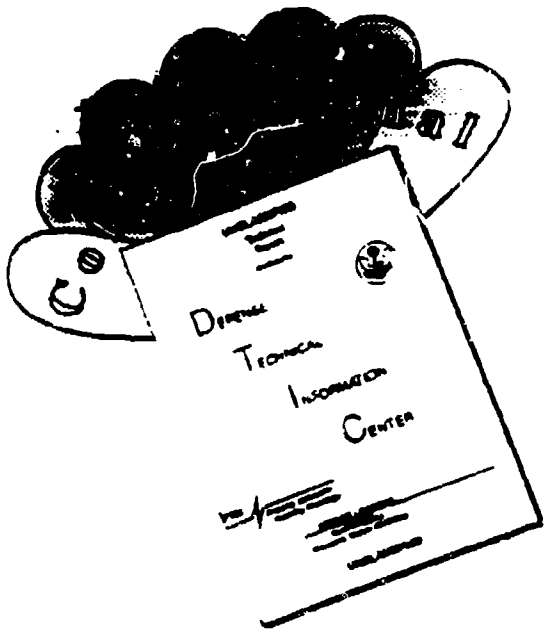

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13. ABSTRACT *(Maximum 200 words)*

A high-pressure windowed strand burner has been used to obtain burning rates for XM46 over the pressure range from 10 to 300 MPa. Burn rates for the liquid XM46 were erratic due to surface irregularities. This burning surface variable was minimized by gelling; two different gelling agents were used. The burn rate (r) vs. pressure (p) can be adequately expressed by an exponential of the form $r = Ap^n$. A definite pressure break occurs around 70 MPa for gelled XM46. For pressures below about 70 MPa, n~0.2 and for pressures between 70 and 300 MPa, the two different gelling agents gave n values of 1.0 and 1.2.

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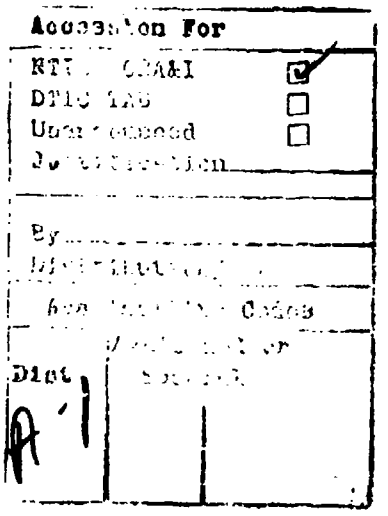

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1. INTRODUCTION

The burning rate of a propellant as a function of pressure is important information for characterizing (modeling) the interior ballistics of a gun. If the pressure exponent for the burning is large ($n > 1$) this can lead (current model predictions [Coffee et al 1989]) to pressure oscillations. In fact, pressure oscillations are of current interest since they have been found to occur in most artillery systems employing XM46 (formerly called LP1846) as a propellant. Oberle and Wren (1990) have determined burn rates for XM46 as a function of pressure in a closed bomb experiment. One of their desires was to obtain viable burn rates without gelling the liquid propellant. Their results indicated a rather large pressure exponent (2.0) for pressures from 100 to 190 MPa. Analysis of their closed bomb data assumed that the propellant regresses in a planar cigarette fashion where the surface area was assumed to remain constant. Previously, McBratney, Bensinger, and Arford (1976) noted substantial surface disturbances on liquid monopropellants for various ignition stimuli and attempted to minimize these irregularities by gelling. The aim of this work is to determine the pressure dependence of the burn rate of a hydroxyl ammonium nitrate (HAN) -based liquid propellant XM46 by photographic observation of the regressing interface. For the ungelled case, surface irregularities were seen for all pressures studied. Gelling XM46 reduced the surface irregularities to a point where an essentially planar burn was established for pressures in the range from about 70 to 300 MPa. For pressures below this range, the regressing gelled surface established a tilted surface as previously observed (McBratney 1980, 1981).

2. EXPERIMENTAL

A windowed steel chamber capable of pressures up to 300 MPa has been used to house liquid propellant samples for photographic studies of their burning characteristics. An illustration of the experiment is shown in Figure 1. The internal diameter of the chamber is 19 cm and the internal volume can be varied from 1 to 6 liters. The liquid propellant samples were typically contained in rectangular acrylic cells with cross-sectional dimensions of 0.3 × 1.0 cm and lengths of about 4 cm. Backlighting of the sample cells was accomplished with a 300-W quartz-tungsten halogen (QTH) lamp. Here the light enters the chamber through a sapphire window of 1.3-cm clear aperture and is subsequently turned 90° by the 45° cut on an acrylic block. Photographic records of the burning behavior were recorded through the 1.27-cm × 5.08-cm rectangular clear aperture of the sapphire observation window by either a half frame 16-mm camera operating at approximately 1,000 frames/s or a 200-frames/s VHS video movie camera. Regression rates were determined over approximately a 2-cm length in the middle portion of the

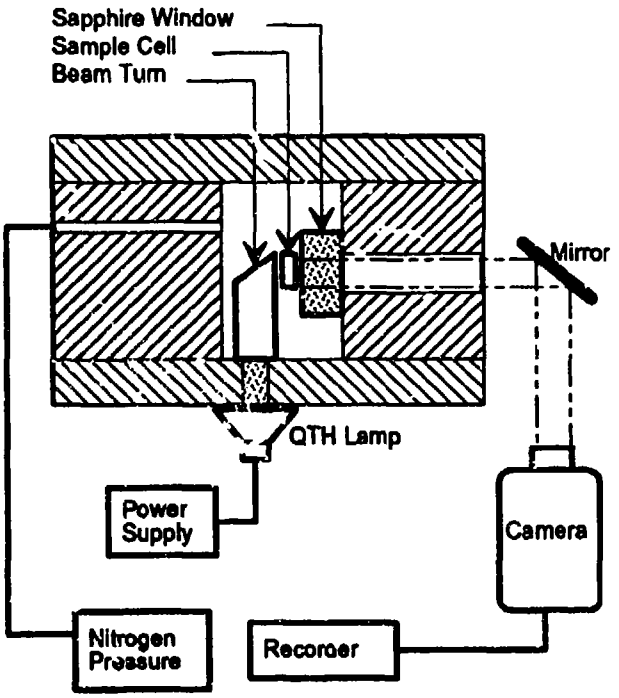

Figure 1. Sketch of the high pressure windowed strand burner (not to scale).

sample cell. Some of the cells had two horizontal marks scribed 2 cm apart on the front surface. All cells had a known length metal needle placed on the back surface of the sample cell. This length provided a calibration for determining burn rates. Moreover, these distinct features residing on both the front and back surfaces of the sample cell provided an opportunity to, in some cases, determine whether the predominant source of light was from the QTH lamp or from combustion occurring within the sample cell.

2.1 Ignition. Ignition of XM46 was accomplished by electrically heating a 0.1-mm-diameter nichrome wire. The bare ignition wire, placed near the top surface of the liquid propellant, created large surface disturbances that were present for the duration of the burn. A number of variations of this technique were tested to see if surface disturbances could be minimized or eliminated. One variation involved coating the ignition wire with a solid mixture consisting of primarily nitrocellulose and black powder. Placement of this coated ignition wire several millimeters above the surface of the liquid propellant created a condition where the liquid propellant would be ignited by the hot combustion gases coming off the coated ignition wire. Surface effects were still seen for this case and it turned out to be an unreliable method of ignition. The variation that has been used for the data obtained in this report involved encasing the ignition wire with gelled XM46 and placing the same on top of the liquid XM46

contained in the sample cell. This method proved to be reliable although varying surface disturbances were still produced and of a sufficient magnitude to be easily observed in the ungelled liquid propellant (see Figure 3).

Burn rates for both ungelled and gelled XM46 have been investigated here and compared with other existing data on similar liquid propellants. These liquid propellant ingredients are given in Table 1.

Table 1. Propellant Mixtures

Liquid Propellant	HAN[a] (weight-percent)	TEAN[b] (weight-percent)	Water (weight-percent)
XM46	60.8	19.2	20.0
LP1845	63.2	20.0	16.8
NOS365	HAN-based		

[a] Hydroxyl ammonium nitrate
[b] Triethanol ammonium nitrate

2.2 Gelling Agents. Several gelling agents were tested with XM46 in order to examine gelling agent effects upon the burn characteristics of gelled propellant. The baseline gelling agent which had been used in earlier HAN propellant tests was Kelzan. This material was originally selected for use with HAN-based propellants by the Naval Ordinance Station at Indian Head (NOSIH) where the Navy liquid propellant formulations were being produced. Kelzan, sold by the Kelco Company, is a gum or polysaccharide obtained from a fermentation process of the bacteria which occurs naturally on plants of the cabbage family. A qualitative description of the gelling capability is that a 50-ml beaker of day-old XM46 mixed with 2% Kelzan can be inverted without movement of material for a time period of minutes.

A fresh sample of Kelzan was obtained from Kelco after it was determined that the available sample was out of date and not functioning as before. However, this newer sample of Kelzan exhibited different behavior when used for gelling XM46. We routinely mix a batch of XM46 with 2% Kelzan and seal this mixture in a clear plastic bag. After several weeks of observation, the mixture (gel) started forming small bubbles and changing color from clear to slightly yellow. When opening the plastic bag an odor similar to vinegar was present. These observations are suggestive that some chemical change is occurring. Contact with the manufacturer of Kelzan was made but no possible reason was uncovered as to why

earlier batches of Kelzan appeared to be unreactive with XM46 while the latest batch exhibited some slow reaction. Thus, another gelling agent was sought. Since Kelzan is a hydrocarbon gelling agent, a nonhydrocarbon gelling agent, Cabosil (silicon dioxide), was tested. Reasonable gels could only be obtained for mixing more than 4% Cabosil in XM46 and we felt that this was too much dilution.

Excellent results have been reported (Giovaenetti 1992) on gelling 13M HAN with Kelco's Welan gum K1A96 at 0.75% gum concentration. We mixed XM46 with 1% Welan gum using a vortex mixing technique. A well-dispersed sample with a slight viscosity increase was produced in a 1-hr setting time. Overnight setting resulted in slightly expanded particles, but very little apparent increase in viscosity. Additionally, the particles had started to settle to the bottom of the container. When testing the sample of Welan gum with tap water, a high viscosity product was obtained. Further testing of Welan gum with XM46 involved heating the mixture to 70° C with vigorous stirring for about 15 min. A large increase in viscosity was obtained, but the sample remained cloudy. Upon cooling to room temperature, the gum had formed an apparent rigid gel, but after overnight storage, the gel had some liquid puddles on the surface. These results were not satisfactory and another Kelco product, Rhamsam gum K1A112, was investigated. This gel is also a fermentation polysaccharide which has been applied to suspension fertilizers. It is compatible with high salt concentration and suspends the components of a suspension fertilizer (e.g., urea, ammonium nitrate and potassium chloride). XM46 was mixed with 1% Rhamsam gum by dribbling the powder slowly into the vortex region formed by a stirrer. Dissolving of the gum was noticed within 15 to 30 min. of stirring with an apparent increase in viscosity. The 24-hr aging period yielded a high viscosity solution, and the windowed chamber burn tests showed well-stabilized burn surfaces. However, over a 6-mo storage time, there is a noticeable decrease in the viscosity of the gel.

3. RESULTS

The regression rate of liquid XM46 (ungelled) vs. pressure is shown in Figure 2 and tabulated in Table 2. Even though the photographic records for these experiments show significant surface irregularities, no attempt has been made to correct for a nonplanar surface. A photo sequence (16-mm camera) showing the regressing surface of ungelled XM46 at 220-MPa nitrogen pressure is shown in Figure 3. The ignition event creates surface disturbances that are present for the duration of the burn and a remnant of the gel encased ignition wire is evident from the brighter emission shown at the top of the left photo. The horizontal lines are scribe marks in the acrylic sample cell spaced 2 cm apart and the vertical line represents a metal pin fixed to the backside of the sample cell. Concave surfaces to the point

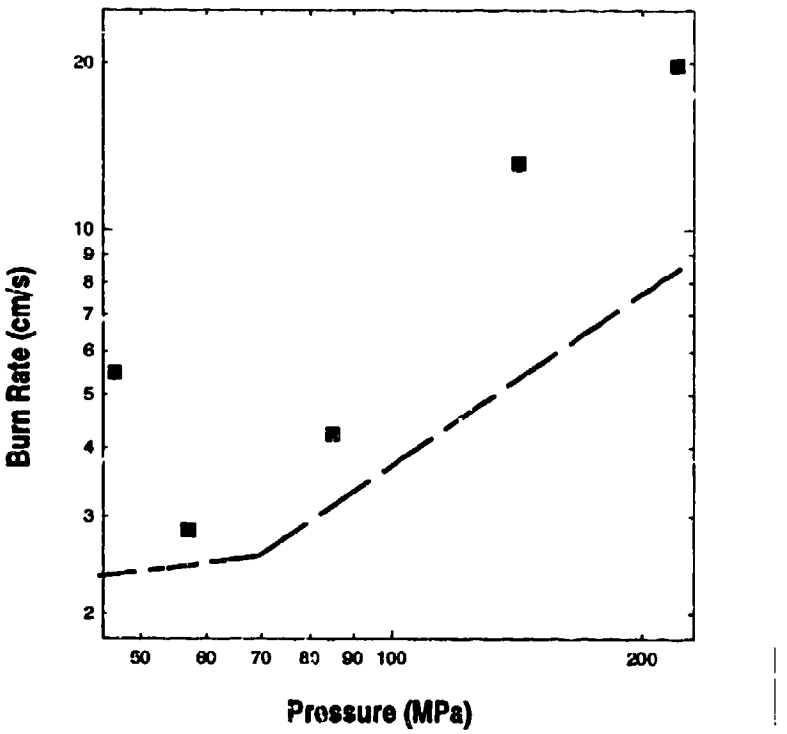

Figure 2. Burn rate vs. pressure for liquid XM46 (■) based on assumption of planar surface. As a contrast, the dashed line represents the gelled XM46 behavior.

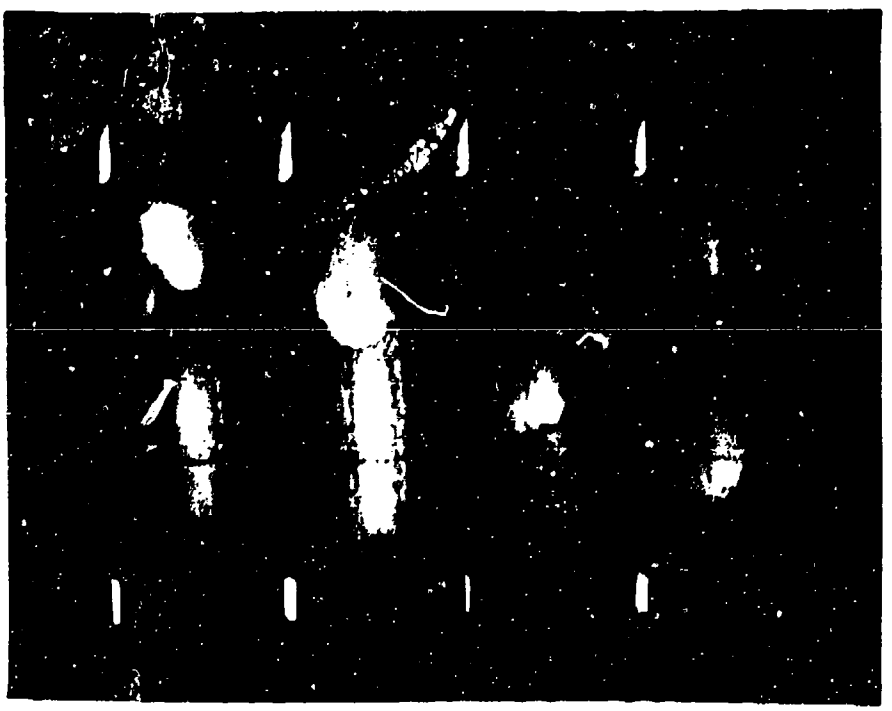

Figure 3. Photo sequence of ungelled XM46 burning at 220-MPa nitrogen pressure. The left is early in the burn just after ignition with the gel encapsulated wire.

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of being semicircular with waves moving on this concave surface are indicated in these and other photo records. Two visually different effects are observed above the surface. There is an increase in brightness (luminosity) where the gas phase begins and there are also regions of reddish-yellow color which originate predominantly from the side walls of the acrylic sample cell at distances of ≥0.5 cm from the liquid surface. Possibly, enhanced heat transfer to these side walls results in some participation of the acrylic in the gas phase combustion. At pressures of 46 and 56 MPa, the surface consists of sloshing waves; however, the concave nature is absent and, as the burn progresses, a slightly convex surface forms. There is an orange region standing off from the surface which could be a flame or nitrogen dioxide emission (see Figure 4). This sequence of photos was obtained from a VHS video camera operating at 200 frames/s.

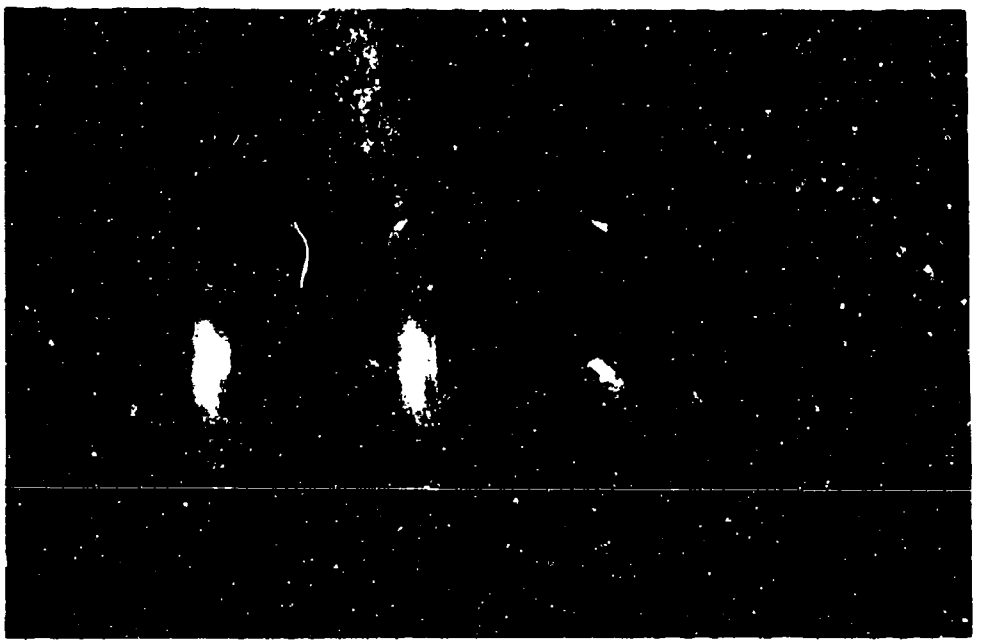

Figure 4. Photo sequence of ungelled XM46 burning at 57-MPa nitrogen pressure.

Selected video frames of gelled XM46 burning surfaces are shown in Figures 5 and 6. The flatness of the gelled surface as a function of nitrogen pressure is illustrated in Figure 5. From left to right the pressures depicted are 296 MPa, 276 MPa, 154 MPa, and 84 MPa. At 84 MPa, the surface appearance

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Figure 5. Video frames of the burning behavior of XM46 gelled with 1% Rhamsam gum. From the left, the nitrogen pressures are 296 MPa, 276 MPa, 154 MPa, and 84 MPa.

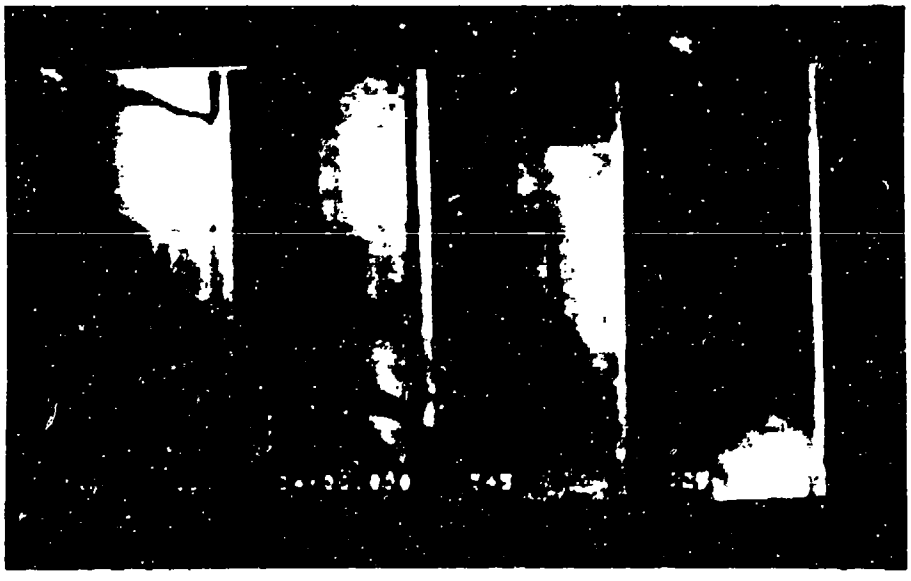

Figure 6. Two-frame sequence of the burning behavior of XM46 gelled with 1% Rhamsam gum. The left two frames are for 34 MPa and the right two for 15.5 MPa.

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is different from the other pressures. Instead of a single reasonably flat line representing the boundary between the gelled solid and the gas phase, two lines can be discerned. For lower pressures, other phenomena appear and are illustrated on Figure 6. The two video frames to the left show behavior at 34 MPa nitrogen. Early in the burn, the surface is fairly flat but develops a convex (cone-like) shape as the burn progresses. The two video frames to the right illustrate the burning behavior of gelled XM46 at 15.5 MPa. In addition to forming the cone shape, a dark residue coating the sample cell walls is left behind.

The regression rate of XM46 gelled with 2% (by weight) Kelzan vs. pressure is shown in Figure 7. The regression rate for XM46 gelled with 1% (by weight) Rhamsam gum is shown in Figure 8 and all of the gelled propellant data are tabulated in Appendix A. Both data sets indicate a break in the pressure dependance around 70 MPa. A least-squares fit to these data using an exponential burn rate law ($r = Ap^n$) as the governing equation gives the following results (r in cm/s).

Pressure range below about 70 MPa

2% Kelzan gelling agent
A = 1.12 ±0.14, n = 0.21 ±0.03
1% Rhamsam gum gelling agent
A = 1.26 ±0.17, n = 0.16 ±0.04

Pressure Range from about 70 MPa to 300 MPa

2% Kelzan gelling agent
A = 0.015 ±0.005, n = 1.18 ±0.06
1% Rhamsam gum gelling agent
A = 0.048 ±0.16, n = 0.96 ±0.06

Data from Figures 7 and 8 are plotted together with data from two other similar HAN-based liquid propellants in Figure 9. All data sets show similar behavior with a pressure break around 70 MPa. The lines drawn in Figures 2, 7, and 8 represent straight-line fits to the data and are not meant to imply that the transition around 70 MPa is that sharp.

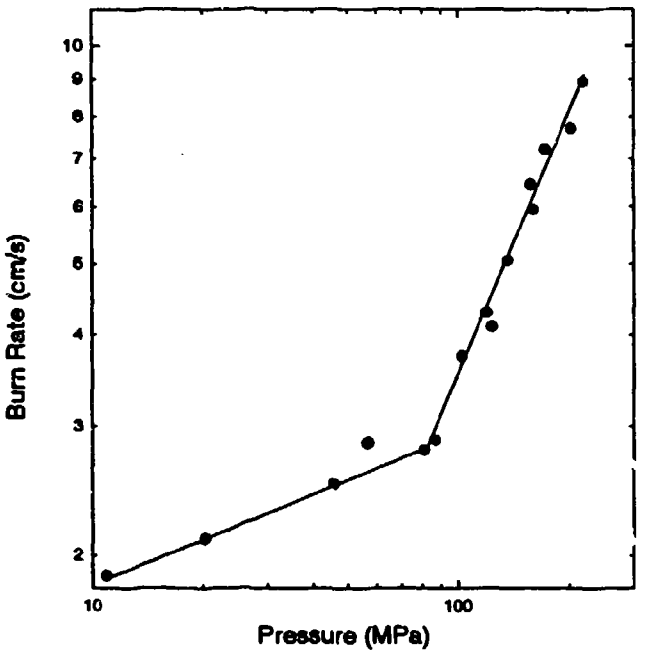

Figure 7. Burn rate of gelled XM46 vs. pressure; gelling agent is 2% Kelzan.

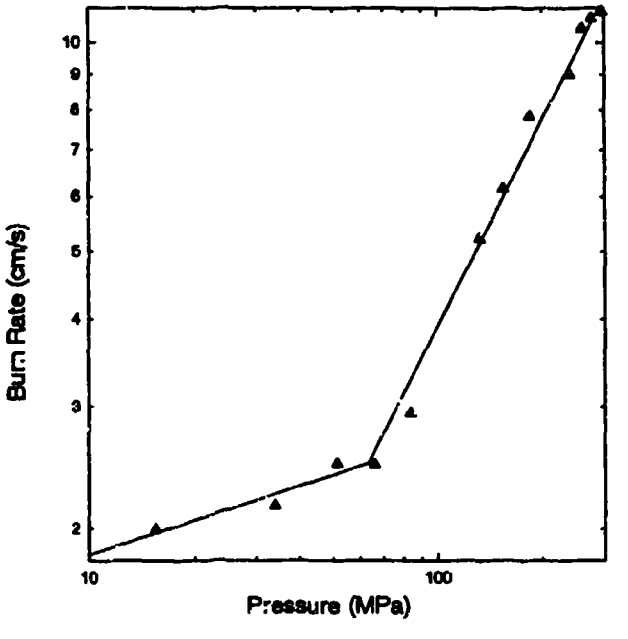

Figure 8. Burn rate of gelled XM46 vs. pressure; gelling agent is 1% Rhamsam.

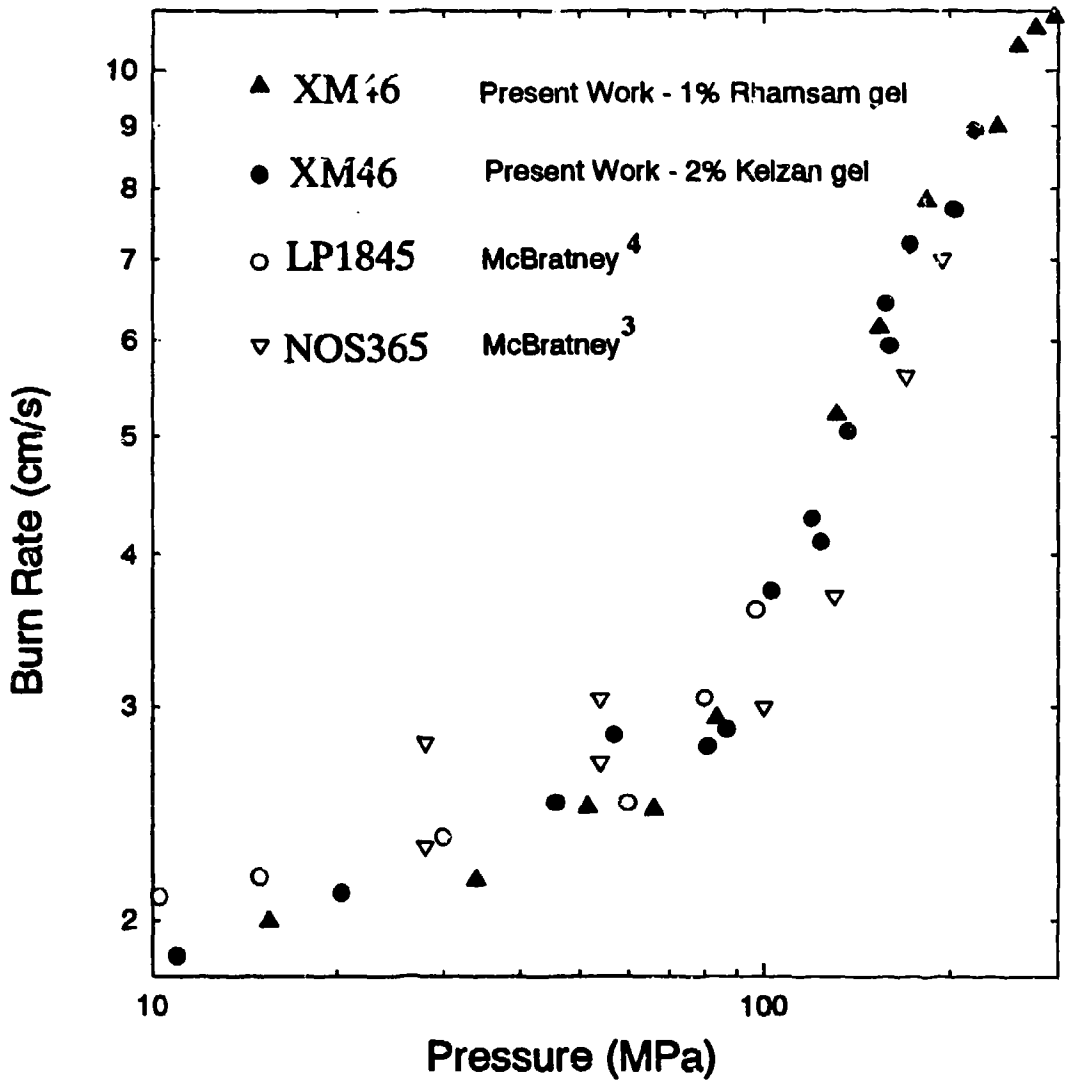

Figure 9. Burn rate data of Figures 7 and 8 plotted together with burn rate results for two other HAN-based gelled liquid monopropellants, LP1845 and NOS-365.

4. DISCUSSION

Consistent trends for liquid propellant burn rates have only been obtained by gelling. In Table 2, there is a comparison between the burn rates obtained for the liquid XM46 versus the nominal burn rates for the gelled XM46. The burn rate ratios vary considerably. Some, or all, of the variability may be removed if a proper surface area could be determined since visual observations readily indicate that the burn surface area is larger than planar for the liquid case. As a qualitative example, assume that the surface generated for the 220-MPa liquid propellant burn (see Figure 3) is semicircular rather than planar. The area increase for this case would be 1.5 and thus lowering the computed burn rate from 20 cm/s to 13 cm/s. For a

cylindrical sample, hemispherical burning involves a surface area twice that of the cross section. The surface area involved here, however, is likely changing as a function of pressure through parameters such as the surface tension (Armstrong and Margolis 1988). Moreover, the ignition stimulus for each experimental run is probably not sufficiently controllable to remove it as a variable property influencing the generated burn surface area.

The last column in Table 2 shows the burn rates computed from closed bomb results of Oberle and Wren 1990, where a pressure exponent of 1.99 was deduced for the high-pressure range. These values are much larger than the values for the gelled propellant and exceed the burn rate measured for the liquid XM46 at the highest pressure.

Table 2. Comparison of Liquid Burn Rates and Nominal Gelled Burn Rates for XM46

Pressure (MPa)	Burn Rate Liquid	Burn Rate Gelled[a]	Burn Rate Ratio	Closed Bomb[b]
46.5	5.5	2.5	2.2	—
57	2.8	2.6	1.1	—
85	4.2	2.9	1.4	4.0
142	13	5.3	2.5	10
220	20	8.9	2.2	25

[a] Values for A and n from the Kelzan gel case given in the text.
[b] Values for A and n given in Oberle and Wren (1990).

The pressure break provides a convenient separation point for discussing possible differences in the combustion mechanisms of gelled XM46. For pressures above about 70 MPa, the regressing surface appears planar with a sharp line defining the burning surface (see Figure 5). At 84-MPa nitrogen pressure, two distinct and separate horizontal lines appear that define the separation between condensed and gas phases. The origin for this feature is not known, but may just be an indication of a tilted surface. At pressures below about 70 MPa, two distinct patterns emerge. In the pressure range from about 20 to 70 MPa, the planar surface develops a convex nature during the course of the burn, but the gas phase region above the surface remains transparent. Below about 20 MPa, the initially planar regressing surface develops inclined surfaces where the gas phase area (dark) above is opaque (see Figure 6). This phenomena has been previously observed by McBratney (1980).

"A probable explanation of the mechanism which causes the convex burn surface at these lower pressures is given by the observation that NOS-365 appears to react in a hypergolic manner with the liquid remnant from the fizz reaction. As this remnant liquid is ejected from the burn front some of it will hit the walls of the test cell and be returned to the reaction area along the cell walls causing a higher reaction rate near the walls."

This explanation is consistent with one of the major conclusions of Vosen (1988), who deduced that the combustion of XM46 is a two-step process. The first step is liquid phase decomposition of HAN, and the subsequent step is the decomposition of TEAN in the gas phase HAN products. The liquid remnant would be droplets of TEAN, and, at sufficiently high pressures, these droplets react. It is not known that the liquid returns to the surface but there could be increased heat transfer to the surface in this region by the boundary condition requirement of zero velocity at the wall. Furthermore, small imperfections along this end wall (vicinity of glue joints) can act as flame holders. Ungelled XM46 forms a concave surface with the sample walls (adhesive property); thus, if the gelled XM46 surface liquifies under combustion conditions, another mechanism for enhanced heat transfer at the walls is present. Any or all of these possible effects could influence planar burning behavior and be pressure dependent.

In their atmospheric pressure studies of LP1845, Zhu and Law (1987) report that propellant explosion is initiated by a liquid phase reaction of the HAN component. Thus, evidence is given for a condensed-phase HAN reaction as being the controlling or rate limiting reaction for XM46 at low pressure. Additional evidence supporting this mechanism is the weak pressure dependence of the burn rate which is characteristic of a controlling condensed phase reaction. At pressures above 70 MPa, the pressure exponent attains values close to 1, which, in the gas phase, is representative of a controlling bimolecular reaction.

As with many experimental research efforts there remains unanswered questions and a few of these are listed below with comments and additional information.

(1) How is the burn rate affected by gelling? This is probably one of the first questions to be asked. What gelling does is to minimize the uncertainty in the burning surface area. Consequently, a better intrinsic burn rate may be determined, although the behavior of the surface is altered from that in a practical combustion situation. Two variables, gel concentration and gel composition,

have been used to determine any significant effects. Kelzan and Rhamsam gum are hydrocarbon gelling agents that have been tested at the 1 and 2% concentration level. Reasonably similar behavior has been observed for both cases. For the high pressure leg, the pressure exponent values were statistically different, however, systematic uncertainty was not taken into account. Attempts were made to test a nonhydrocarbon gelling agent, Cabosil, but unacceptably high gel concentrations were required to sufficiently increase the viscosity.

(2) Does the sample cell geometry affect the burn rate? Since the burn rate measurements for all three gelled HAN-based liquid propellants are in good agreement and the sample cells were of different geometry (see Appendix B), this effect is probably minimal. Moreover, for the gelled case, the influence of sample cell geometry is reduced since surface disturbances have been minimized. Nonetheless, heat losses to the walls is still present as a variable.

(3) Does the sample cell material affect the burn rate? The majority of experiments were done with an acrylic sample cell. This material has a low thermal conductivity; thus, less heat is carried away than for most other common cell materials (i.e., the thermal conductivity for acrylic is about a factor of 8 lower than quartz). However, temperatures may be attained such that a chemical reaction could occur between the combusting propellant and the acrylic. Several experiments were conducted where a quartz cell of similar geometry was used to contain the Kelzan gelled XM46 sample. The results of these measurements at 73 and 91 MPa gave burn rates of 3.2 and 3.4 cm/s, respectively. These values are about 15% and 8% higher than the average values obtained from fitting the data of Figure 7. These results, in the context of estimated uncertainty for the measurements, suggest that the cell material is not significantly affecting the burn rate measurements.

(4) What drives the pressure break in the burn rate of the gelled HAN-based liquid propellants? Several guesses for this phenomenon are offered.

a. The condensed phase temperature gradient increases with pressure to a point where the controlling reaction operates at the surface or the gas phase.

b. As the pressure increases the gas phase heat release reactions approach the surface thus allowing for increased heat feedback to the surface.

c. Condensed phase components are ejected from the regressing surface at lower pressure and the pressure break region is where these components volatilize at the surface.

d. This is a manifestation of critical point behavior. Since the pressure break appears reasonably distinct, changes in properties associated with reaching the critical point drive the change in the pressure dependence. Kounalakis and Faeth's (1988) calculations suggest an unusually high value of 250 MPa ±125 MPa for the critical combustion pressure. This value is most sensitive to the interaction parameter for TEAN and water due to low volatility of TEAN. A low estimate of around 60 MPa for the critical combustion pressure is also calculated when the effect of this interaction parameter is minimized (Kounalakis and Faeth 1988).

(5) Burn rates of XM46 are reported here for two different gelling agents, Kelzan and Rhamsam. For pressures above about 70 MPa the differences in the determined pressure exponent are larger than the one standard deviation uncertainties would suggest. Only the Rhamsam data have been extended to pressures in excess of 220 MPa, thus the three highest pressure points were removed from the Rhamsam data case. This reduced data set was refitted over the range from 70 to 239 MPa. The fit gives $A = 0.012 \pm 0.002$ and $n = 1.24 \pm 0.03$ in much closer agreement with the Kelzan data. Perhaps there is another slope break above 200 MPa.

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APPENDIX A:

GELLED LIQUID PROPELLANT BURN RATE DATA

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Table A-1. Gelled Liquid Propellant Burn Rate Data as a Function of Pressure

Pressure (MPa)	XM46 Burn Rate - 2% Kelzan gel (cm/s)	XM46 Burn Rate - 1% Rhamsam gum (cm/s)	LP1845 Burn Rate - 2% Kelzan gel (cm/s)	NOS365 Burn Rate - 2% Kelzan gel (cm/s)
10.3	—	—	2.09	—
11.0	1.87	—	—	—
15.0	—	—	2.17	—
15.5	—	2.00	—	—
20.4	2.10	—	—	—
28.0	—	—	—	2.30
28.0	—	—	—	2.80
30.0	—	—	2.34	—
34.0	—	2.16	—	—
46.0	2.50	—	—	—
51.5	—	2.48	—	—
54.0	—	—	—	2.70
54.0	—	—	—	3.05
57.0	2.84	—	—	—
60.0	—	—	2.50	—
66.0	—	2.47	—	—
80.0	—	—	3.05	—
81.0	2.78	—	—	—
83.5	—	2.94	—	—
87.0	2.87	—	—	—
97.0	—	—	3.60	—
100	—	—	—	3.00
103	3.73	—	—	—
120	4.28	—	—	—
124	4.09	—	—	—
130	—	—	—	3.70
132	—	5.21	—	—

Table A-1. Gelled Liquid Propellant Burn Rate Data as a Function of Pressure (continued)

Pressure (MPa)	XM46 Burn Rate - 2% Kelzan gel (cm/s)	XM46 Burn Rate - 1% Rhamsam gum (cm/s)	LP1845 Burn Rate - 2% Kelzan gel (cm/s)	NOS365 Burn Rate - 2% Kelzan gel (cm/s)
137	5.04	—	—	—
154	—	6.15	—	—
158	6.43	—	—	—
160	5.93	—	—	—
170	—	—	—	5.60
173	7.20	—		—
184	—	7.82	—	—
195	—	—	—	7.00
204	7.68	—	—	—
220	8.92	—	—	—
239	—	8.99		—
259	—	10.5	—	—
276	—	10.8	—	—
296	—	11.1	—	—

APPENDIX B:

SAMPLE CELL GEOMETRY

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Table B-1. Sample Cell Geometry

Gelled Liquid Propellant	Cross Section	Dimensions	Material	Reference
XM46 - 2% Kelzan gel	Rectangular	0.3 × 1.0 cm	Acrylic	This report
XM46 - 1% Rhamsam gel	Rectangular	0.3 × 1.0 cm	Acrylic	This report
LP1845 - 2% Kelzan gel	Rectangular	0.15 × 0.6 cm	Acrylic	McBratney (1981)
NOS365 - 2% Kelzan gel	Rectangular	0.15 × 0.6 cm	Acrylic	McBratney (1980)
NOS365 - 2% Kelzan gel	Tube	0.38 cm dia.	Polypropylene	McBratney (1980)

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